

# PATENT SPECIFICATION

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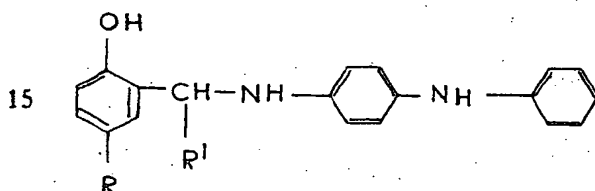


## (54) SUBSTITUTED *p*-PHENYLENEDIAMINES

(71) We, KODAK LIMITED, a Company registered under the Law of England, of Kodak House, Station Road, Hemel Hempstead, Hertfordshire, do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

This invention relates to new *N*-(*o*-hydroxybenzyl) - *N'* - phenyl - *p* - phenylenediamines, and to a method for their preparation.

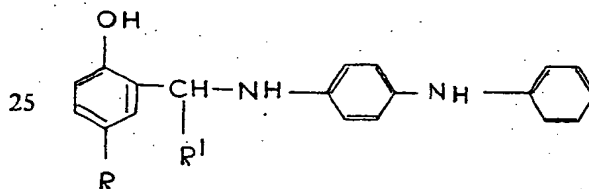
In accordance with the present invention, there is provided a compound having the formula:



wherein R is an alkyl group having from 1 to 20 carbon atoms and R<sup>1</sup> is an alkyl group having from 1 to 20 carbon atoms, or a phenyl group.

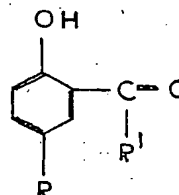
20 Compounds in accordance with the invention are useful as antidegradants for rubber.

Also in accordance with the present invention, there is provided a method of preparing a compound having the formula:



wherein R is an alkyl group having from 1 to 20 carbon atoms and R<sup>1</sup> is an alkyl group

having from 1 to 20 carbon atoms, or a phenyl group, the method comprising forming a Schiff's base by condensing *N*-phenyl-*p*-phenylenediamine with a compound having the formula



in the presence of piperidine, and catalytically hydrogenating the Schiff's base.

The use of piperidine as a catalyst in the above method gives almost quantitative yields of the Schiff's base.

In order that the invention may be more clearly understood, Examples thereof will now be given. In all the Examples the procedure was as follows:

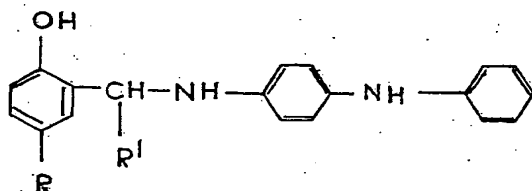
*N* - phenyl - *p* - phenylenediamine (0.125 mole), a 2 - hydroxy - 5 - alkyl - phenylalkylketone (0.1375 mole), toluene (4 ml) and piperidine (8 ml) were heated at 170°C for 7 hrs. The water of reaction was removed using a Dean and Stark head. The reaction solvent was evaporated, the Schiff's base was dissolved in acetone (500 ml) and 10% platinum on charcoal catalyst (1.5 g) added. Hydrogenation at 100 p.s.i. hydrogen and 50°C gave the *N* - (*o* - hydroxybenzyl) - *N'* - phenyl - *p* - phenylenediamine as an oily residue which crystallised either from petroleum ether or a mixture of petroleum ether and ethyl acetate.

The ketone used in each Example is indicated by the values of R and R<sup>1</sup> given in the following Table, which also shows the melting point of the final product and the yield.

Ex.	R	R <sup>1</sup>	Product	mp. °C	% Yield
1	CH <sub>3</sub>	C <sub>2</sub> H <sub>5</sub>	N-(2-hydroxy-5-methyl-2-ethyl-benzyl)-N'-phenyl- <i>p</i> -phenylene-diamine	114-116	49
2	CH <sub>3</sub>	<i>n</i> -C <sub>3</sub> H <sub>7</sub>	N-(2-hydroxy-5-methyl- $\alpha$ -propyl-benzyl)-N'-phenyl- <i>p</i> -phenylene-diamine	118-119	62
3	CH <sub>3</sub>	CH <sub>3</sub>	N-(2-hydroxy-5-methyl- $\alpha$ -methyl-benzyl)-N'-phenyl- <i>p</i> -phenylene-diamine	89-90	50
4	C <sub>2</sub> H <sub>5</sub>	C <sub>2</sub> H <sub>5</sub>	N-(2-hydroxy-5-ethyl- $\alpha$ -ethyl-benzyl)-N'-phenyl- <i>p</i> -phenylene-diamine	90-92	41
5	CH <sub>3</sub>	C <sub>6</sub> H <sub>5</sub>	N-(2-hydroxy-5-methyl- $\alpha$ -phenyl-benzyl)-N'-phenyl- <i>p</i> -phenylene-diamine	144-145	55

WHAT WE CLAIM IS:—

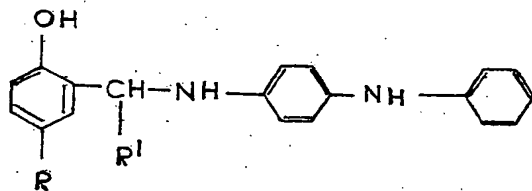
1. A compound having the formula:



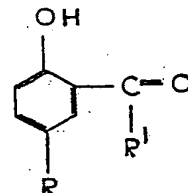
5 wherein R is an alkyl group having from 1 to 20 carbon atoms and R<sup>1</sup> is an alkyl group having from 1 to 20 carbon atoms, or a phenyl group.

10 2. A compound as claimed in Claim 1 and as specified in any one of Examples 1 to 5 herein.

3. A method of preparing a compound having the formula:



wherein R is an alkyl group having from 1 to 20 carbon atoms and R<sup>1</sup> is an alkyl group having from 1 to 20 carbon atoms, or a phenyl group, the method comprising forming a Schiff's base by condensing N - phenyl - *p*-phenylenediamine with a compound having the formula:



in the presence of piperidine, and catalytically hydrogenating the Schiff's base.

4. A method as claimed in Claim 3 and substantially as hereinbefore described in any one of Examples 1 to 5.

5. A compound as claimed in Claim 1 whenever prepared by a method as claimed in Claim 3.

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